

Theory of isotope effect in photoemission spectra of high- T_c superconducting cuprates

G. Seibold¹ and M. Grilli²

¹*Institut für Physik, BTU Cottbus, PBox 101344, 03013 Cottbus, Germany and*

²*Istituto Nazionale di Fisica della Materia, Unità Roma 1 and SMC Center, and Dipartimento di Fisica
Università di Roma "La Sapienza" piazzale Aldo Moro 5, I-00185 Roma, Italy*

(Dated: February 2, 2008)

We investigate the effect of isotope substitution on the electronic spectral functions within a model where the charge carriers are coupled to bosonic charge-order (CO) fluctuations centered around some mean frequency ω_0 and with enhanced scattering at wave-vector \mathbf{q}_c . It is shown that a mass dependence of ω_0 is not sufficient in order to account, especially at high energies, for the dispersion shifts experimentally observed in an optimally doped superconducting cuprate. We argue that isotope substitution induces a change of the spatial CO correlations which gives good account of the experimental data.

PACS numbers: 74.72.-h, 74.20.Mn, 71.38.Cn, 79.60.-i

Several isotope effects (IE) have been found in underdoped superconducting cuprates, not only in the critical temperature but also in the superfluid density, in the fermion effective mass, and in the dynamic lattice distortion [1] and in the pseudogap crossover temperature T^* [2]. More recently a remarkable IE has also been found in angle-resolved photoemission spectroscopy (ARPES) experiments on optimally doped Bi-2212 [3]. In this experiment the dispersive single-particle excitations were investigated and strong peculiar variations upon replacing ^{16}O with ^{18}O were found. Specifically, little isotopic dependence was found both for the kink and the low-energy part of the spectra along the $\Gamma \rightarrow X$, [i.e., $(0,0) \rightarrow (\pi,\pi)$] direction. On the other hand the dispersive “hump” at higher binding energy displayed a sizable IE, which caused a shift of about 15 meV towards larger binding energies. Around the M points at $(\pi,0)$ and $(0,\pi)$ the IE has a reversed sign and both the low- and high-energy parts of the dispersion shift to smaller binding energies upon isotope substitution. In this region shifts as large as 40 meV are reported. This IE is absolutely non trivial in various respects. First of all, the ion replacement is expected to change by only 6% the phonon frequencies involving oxygen ions. The changes of the electron dispersions are large and within a standard perturbative Eliashberg-like scheme would imply too large e-ph couplings [4]. Secondly the isotopic shifts have different signs depending on the region of the Fermi surface. Thirdly they do not substantially occur around the “kinks” of the dispersion curves, as it would be expected if the isotope replacement were only changing the frequency of the phonon [or more generally of the collective mode (CM)] interacting with the quasiparticles (QP’s).

In this letter we specifically address the issue of this peculiar IE in ARPES and we provide a natural explanation for it within the theory based on the occurrence of a charge-ordering (CO) quantum-critical point (QCP) around optimal doping of the cuprates [5]. This theory predicts the occurrence of a CO phase in a substantial

region of the phase diagram below a doping-dependent critical temperature $T_{CO}(x)$ ending at zero temperature in a QCP around optimal doping. Actually several mechanisms (pairing, disorder, and low dimensionality) may prevent a fully realized long-range CO state. Nevertheless, despite this “missed” criticality, in the low-temperature underdoped region below $T^* \sim T_{CO}$, local dynamical CO should be revealed as a natural inclination of the system. After this theoretical prediction, many evidences have arisen that a QCP could be hidden under the superconducting “dome” [6, 7]. The prediction of a local CO in the underdoped cuprates is also finding new experimental confirmations [8], which corroborate less recent observations of “stripe” phases. Within the CO-QCP theory it was also possible to explain the peculiar IE of the pseudogap crossover temperature T^* mentioned above [9]. Therefore it seems legitimate to adopt this theoretical framework to address the issue of IE in ARPES experiments. In particular there are several characteristics of the theory, which render it a particularly suitable scheme: a) the presence of a “missed” criticality justifies the presence of low-energy (nearly critical) charge collective modes (CM), which have already been shown to produce “kinks” in the dispersive spectra [10]; b) on general grounds, the proximity to a (“missed”) criticality renders the system highly susceptible so that large effects (e.g. large IE on the single-particle excitations) can arise from small changes (like a 6% shift of some phonon frequencies); c) within the Hubbard-Holstein model, which was the starting point of the CO-QCP theory, the CM are mixed phonon-plasmon modes, which therefore have both a built-in dependence on the phonon frequency naturally leading to IE’s and an electronic character, which renders them similar to the spin modes considered in Refs.[11]; d) the CM’s are only critical near specific wavevectors \mathbf{q}_c related to the wavelengths of the CO textures. As a consequence they have a strong momentum dependence, which allows them to play different roles in interacting with QP’s in different regions of the Brillouin zone. As

we shall see this is a crucial feature in order to explain the opposite sign of the IE on dispersive bands along the nodal and antinodal directions. Within the above framework we performed a systematic analysis of the effects of changing the CM frequency ω_0 , the QP-CM coupling g , and the CO inverse correlation length γ . Our main finding is that changes of ω_0 and/or g alone by no means allow to reproduce the experimental results. On the other hand the crucial parameter to vary in order to semiquantitatively reproduce the experimental isotopic shift is the inverse charge-charge coherence length γ . Before reporting on our detailed calculations, we first present qualitative arguments to give an intuitive understanding of this result. We start, e.g., from an hole state at wavevector \mathbf{p} , which is scattered by a narrow CM of energy ω_0 and wavevector \mathbf{q}_c . This hole acquires a self-energy $\Sigma_p(\omega) = g^2/(\omega + \omega_0 - \varepsilon_{\mathbf{p}+\mathbf{q}_c})$. If the CM has instead a wavevector broadening γ , the self-energy becomes proportional to the hole density of states (DOS) $\rho_{\mathbf{p}+\mathbf{q}_c}$ in the region of the momenta, where the hole is scattered by the CM. In the two-dimensional case of our interest this region has an area γ^2 around $\mathbf{p} + \mathbf{q}_c$. If the DOS does not vary appreciably in the Brillouin zone, one obtains a result similar to the one found by Engelsberg-Schrieffer [12] which was derived under the assumption of electron-hole symmetry and a q-independent matrix element between the electrons and the CM (a Holstein phonon): There is a dispersion break at a binding energy $\omega \sim \omega_0$ and a reduction of ω_0 always leads to a shift of the electron dispersion at higher binding energies (and only in a limited energy range around the break). On the other hand, if $\rho_{\mathbf{p}+\mathbf{q}_c}$ varies substantially around the Fermi surface, a different situation can occur. Let us suppose that the isotope replacement acts on γ [e.g., $\gamma(^{18}\text{O}) > \gamma(^{16}\text{O})$, this is what we find to be the relevant effect in our detailed calculations]. Then upon isotopic substitution the hole can be scattered in a broader momentum region where the average DOS can be substantially different. In particular, if the local DOS increases $\text{Re}\Sigma(^{16}\text{O}, \omega < -\omega_0) < \text{Re}\Sigma(^{18}\text{O}, \omega < -\omega_0)$ (cf., e.g., Fig. 2c) and as a result the electronic dispersion is shifted upwards to lower binding energies $\varepsilon_k(^{18}\text{O}) > \varepsilon_k(^{16}\text{O})$. This is indeed the desired effect to explain the experiments of Ref. [3] near the M points.

In order to substantiate these ideas within more detailed calculations we consider a system of superconducting electrons exposed to an effective action

$$S = -g^2 \sum_{\mathbf{q}} \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \chi_{\mathbf{q}}(\tau_1 - \tau_2) \rho_{\mathbf{q}}(\tau_1) \rho_{-\mathbf{q}}(\tau_2) \quad (1)$$

describing dynamical incommensurate CO fluctuations. Using Nambu-Gorkov notation the unperturbed Matsubara Green's functions $\underline{\underline{G}}^0$ are 2×2 matrices and the leading order one-loop contribution to the self-energy reads

as

$$\underline{\underline{\Sigma}}(\mathbf{k}, i\omega) = -\frac{g^2}{\beta} \sum_{\mathbf{q}, ip} \chi_{\mathbf{q}}(ip) \underline{\underline{\tau}}_z \underline{\underline{G}}^0(\mathbf{k} - \mathbf{q}, i\omega - ip) \underline{\underline{\tau}}_z \quad (2)$$

which in turn allows for the calculation of $\underline{\underline{G}} = \underline{\underline{G}}^0 + \underline{\underline{G}}^0 \underline{\underline{\Sigma}} \underline{\underline{G}}$. The spectral function can be extracted from $\underline{\underline{A}}_{\mathbf{k}}(\omega) = (1/\pi) \text{Im} \underline{\underline{G}}_{11}(\mathbf{k}, \omega)$.

In order to simplify the calculations we consider a Kampf-Schrieffer-type model susceptibility [16] which is factorized into an ω - and q-dependent part, i.e.

$$\chi_{\mathbf{q}}(i\omega) = W(i\omega) J(\mathbf{q}) \quad (3)$$

where, choosing a unit lattice spacing,

$$J(\mathbf{q}) = \frac{\mathcal{N}}{4} \sum_{\pm q_x^c; \pm q_y^c} \frac{\gamma}{\gamma^2 + 2 - \cos(q_x - q_x^c) - \cos(q_y - q_y^c)}. \quad (4)$$

\mathcal{N} is a suitable normalization factor introduced to keep the total scattering strength constant while varying γ . The (real-frequency)-dependent part $W(\omega) = \int d\nu F_{\omega_0, \Gamma}(\nu) 2\nu/(\omega^2 - \nu^2)$ with $F_{\omega_0, \Gamma}(\omega) \sim \Gamma/[(\omega - \omega_0)^2 + \Gamma^2]$ is a normalized lorentzian distribution function centered around ω_0 with halfwidth Γ . If only this part were present in $\chi(i\omega)$, one would have a bosonic spectrum $B(\omega) = \tanh(\omega/(kT)) F_{\omega_0, \Gamma}(\omega)$ which is a “smeared” version of the Holstein phonon considered in Ref.[12]. The crucial feature of the susceptibility (3) is the substantial momentum dependence, which describes the local order formation and reflects the proximity to a (missed) CO instability. This susceptibility contains the charge-charge correlations which are enhanced at the four equivalent critical wave vectors $(\pm\pi/2, 0)$, $(0, \pm\pi/2)$. The static limit $g(\nu) = \delta(\nu)$ together with an infinite charge-charge correlation length $\gamma \rightarrow 0$ reproduces mean-field results for a long-range CO phase [17]. For the bare electron dispersion used in the following

$$\varepsilon_k = -2t(\cos(k_x) + \cos(k_y)) + 4t' \cos(k_x) \cos(k_y) - u$$

the parameters have been fitted so that the resulting spectral functions approximately agree in energy with the data of Ref. [3]. We use $t = 0.35\text{eV}$, $t'/t = 0.32$, $u = -0.42\text{eV}$. The superconducting d-wave gap is $\Delta_k = 35\text{meV}[\cos(k_x) - \cos(k_y)]/2$. The width of the lorentzian in frequency space is $\Gamma = 50\text{meV}$

In Ref. [17] we demonstrated that a static two-dimensional eggbox-type charge modulation with \mathbf{q}_c oriented along the (1,0), (0,1) directions can account for the basic Fermi surface (FS) features in the optimally and underdoped Bi2212 compounds. In a subsequent paper [10] we analyzed ARPES spectra in Bi-2212 showing that the main dispersive features [18, 19] were reproduced, including the well-known kink in the nodal dispersion which appears as a break for momentum scans far from the node [19]. The main point to be recollected here is that the

M-points define the “hot” regions for incommensurate CO scattering. In these regions of k -space the low-energy QP’s are strongly scattered by the CM’s into other states with high DOS (still near the M points). On the other hand, along $\Gamma - X$ the coupling of electronic states to the CO mode is significantly reduced (“cold” region) at low binding energies and one finds weakly interacting QP’s which get heavily damped only at high binding energies. Here the QP’s both those at low and at high binding energies are scattered by the CM’s into regions with rather low DOS. This different character between “hot” and the “cold” regions is a key point to understand the behavior of the ARPES-IE, with the “surprising” change of sign moving from diagonal scans to scans near M and sets the stage for our analysis.

— *Only mode-frequency change* — First of all we tried a simple change of the CM frequency only. Since we are not dealing with simple phonons, but rather with (nearly critical) CM’s, it is difficult to *a priori* determine the isotopic shift (if any) of the CM frequency. We take $\omega_0(^{16}\text{O}) = 75\text{meV}$, $\omega_0(^{18}\text{O}) = 60\text{meV}$ to optimize the agreement with experiments. The frequency shift is quite large (15meV) with respect to the one expected for phonons. The coupling constant is $g(^{16}\text{O}) = g(^{18}\text{O}) = 0.13\text{eV}$, which leads to self-energies at the nodal point of the same order of magnitude than those reported in [3]. Both along the diagonal direction and along the vertical cut corresponding to cut #7 in Ref. [3] there is a significant isotope shift only in a narrow energy window near the kink around -0.1eV . Our dispersions do not display an isotope shift in the high energy region. Upon changing in the opposite way the frequency $\omega_0(^{16}\text{O}) < \omega_0(^{18}\text{O})$ the agreement with experiments is even worse. Therefore, within our model and despite the large shift in ω_0 the IE can not be ascribed to a simple shift of the CM frequency.

— *Coupling and mode-frequency change* — As before we choose $\omega_0(^{16}\text{O}) = 75\text{meV}$ and $\omega_0(^{18}\text{O}) = 60\text{meV}$. In addition we consider now different coupling constants for the isotopes. g can be determined from the requirement that the normal contribution to the self-energy $\Sigma_{11}(k_F, E_F)$ at the nodal point gives a reasonable fit to the $\text{Re}\Sigma$ extracted in Ref. [3]. We find $g(^{16}\text{O}) = 0.13\text{eV}$ and $g(^{18}\text{O}) = 0.114\text{eV}$. Along the diagonal direction one finds qualitatively the same behavior as with ω_0 changes only: the IE is still confined to a rather narrow energy window. Moreover the theoretical dispersions along the vertical cut do not properly describe the findings of Ref.[3]: contrary to the experiments, with isotopic replacement the low-energy part of the dispersion for ^{18}O is shifted at higher energies with respect to the dispersion for ^{16}O . Furthermore, while ^{16}O shows the break in the dispersion as in the experiment, the lower coupling for ^{18}O turns this break into a kink.

— *Only change of γ* — Finally we consider a change of γ alone. We take $\omega_0(^{16}\text{O}) = \omega_0(^{18}\text{O}) = 50\text{meV}$ and

$g(^{16}\text{O}) = g(^{18}\text{O}) = 0.16\text{eV}$, but we choose different spatial correlation lengths, i.e. $\gamma = 0.15$ for ^{16}O and $\gamma = 0.30$ for ^{18}O . Fig. 1 shows energy distribution curves (EDC’s) and the corresponding dispersions for a cut along the diagonal (Fig. 1 upper panel) and a vertical cut near the M-point similar to cut #7 in Ref. [3] (Fig. 1 lower panel, cf. inset to Fig. 2d). Along the diagonal direction, below the kink energy we find a shift of the ^{18}O dispersion towards larger binding energies. Similarly to what is experimentally observed, the IE shift extends far below the kink energy. In the low energy regime above the kink the shift becomes negligible like in the experiments. One only finds a minor discrepancy with experiment since in the crossover region the theoretical ^{16}O dispersion falls below the ^{18}O one. In the vertical cut

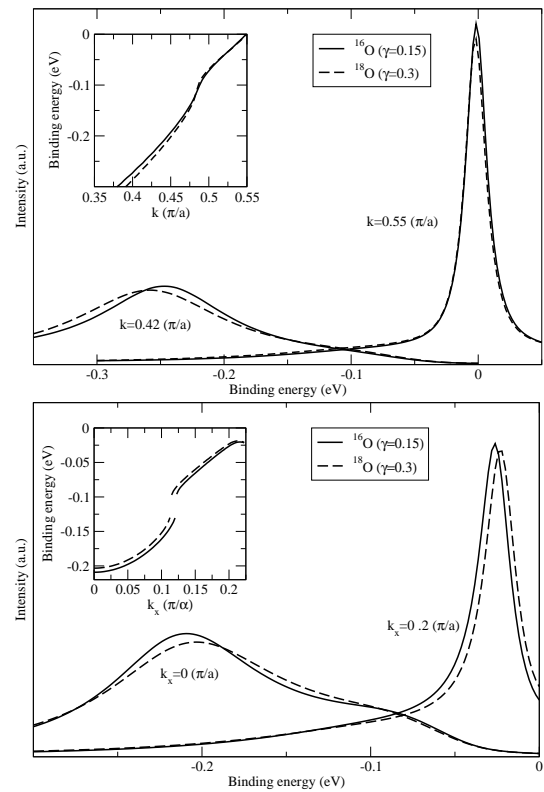


FIG. 1: EDC’s and the corresponding EDC-derived dispersions (inset) for the case where only γ is varied upon isotope substitution. Intensities have *not* been scaled to the same amplitudes as done in Ref. [3]. Upper panel: diagonal cut; Lower panel: vertical cut at $k_y = 0.6(\pi/a)$ corresponding to cut 7 in Fig. 2 of Ref. [3] (cf. inset to Fig. 2d).

as shown in Fig. 1 (lower panel) one sees that the ^{18}O dispersion is shifted *above* the ^{16}O dispersion both at low and high binding energies. This behavior is also in agreement with the experimental findings of Ref. [3]. We notice that, despite the qualitative agreement, the calculated shift ($\sim 15 - 20\text{meV}$) is substantially smaller than the experimental one ($\sim 30 - 40\text{meV}$). However, this quantitative discrepancy could be solved by assuming a

(more realistic) momentum dependence of the QP-CM mode coupling g , which instead we take constant for simplicity reasons.

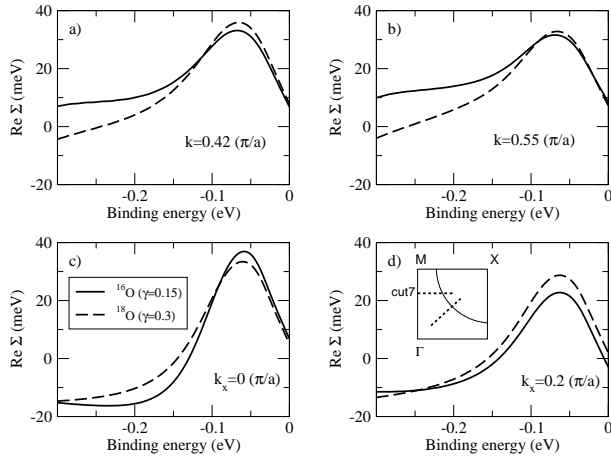


FIG. 2: Real part of the normal contribution to the self-energy $Re\Sigma_{11}(q, \omega)$. Panels (a,b) show $Re\Sigma_{11}$ for two momenta along the nodal direction and panels (b,c) are for two momenta on cut #7 (cf. inset to d).

Fig. 2 shows $Re\Sigma_{11}(q, \omega)$ for the case where γ is increased by isotope substitution, while keeping ω_0 and g fixed. Results for two momenta along diagonal cuts are reported in the two top panels, where the most prominent feature is that at large binding energies the self-energy generically decreases upon increasing γ (i.e. upon decreasing the CO correlation length). Remarkably this reduction persists down to very high binding energies. While this reduction is expected on general grounds even for the phonon case, [4], the frequency extension of this effect is rather unusual and arises from the plateau in the high-energy part of the self-energy occurring when γ is small. On the other hand the self-energy generically varies little at low energies, while a moderate increase, $Re\Sigma(^{18}O) > Re\Sigma(^{16}O)$, occurs near its maximum (around which the kink in the dispersion takes place) due to the enhancement of the phase space of the scattered QP states induced by the increased γ [21]. This effect is even more pronounced in the antinodal region reported in the two lower panels. Along cut #7 one therefore observes a shift of the ^{18}O dispersion to smaller binding energies.

— *Conclusions* — We applied here a simple perturbative scheme of superconducting quasiparticles coupled to charge CM’s. From the above systematic analysis we find that, if the isotopic substitution changes the CM momentum broadening γ , our model accounts both for the large energy range in which isotopic shifts are observed and for the change in sign of these shifts moving from the $\Gamma \rightarrow X$ cuts to the cuts near the M points. The fact that the crucial effect of the isotopic replacement is to change the CO coherence length (inversely related

to γ) also supports the idea that charge fluctuations are highly correlated in optimally doped cuprates. This is consistent with Ref. [9], where it was found that the $^{16}O \rightarrow ^{18}O$ replacement shifts the (“missed”) quantum-critical line to higher dopings (or equivalently confers to the system properties of more underdoped materials). For optimally or underdoped materials at low temperature this “pushes” the system more deeply in the nearly ordered region away from the “missed” critical line, naturally reducing the spatial CO correlations. An additional broadening of the CM momentum distribution can arise from disorder, if the $^{16}O \rightarrow ^{18}O$ substitution is incomplete. Another possible way of arguing can be derived from Gutzwiller+RPA calculations of the stripe phase, which indicate that upon underdoping the intensity distribution of the mode becomes more extended in momentum space [22]. All these arguments provide support to an increase of γ upon $^{16}O \rightarrow ^{18}O$ substitution.

Since it turns out that isotopic changes of ω_0 and of g play a minor role, one could argue that the CM’s not necessarily are charge modes (mixed with phonons), but could as well be spin modes. Indeed, our perturbative scheme previously adopted in Ref. [10] provided an interpretation of spectral features in Bi-2212 quite similar to those of Ref. [11], where the coupling of electrons to antiferromagnetic (AF) spin CM’s was considered. This is natural because for both AF and CO scattering the states near the M points are the “hot spots”, i.e. are most strongly affected by the scattering. To (at least preliminarily) discriminate between charge and spin modes, we also checked whether CM’s peaked around $\mathbf{Q}_{AF} = (\pi, \pi)$ relevant for AF fluctuations could account for the IE in ARPES. While a more detailed analysis of AF modes is beyond the scope of this work, we found that CM’s peaked at \mathbf{Q}_{AF} fail in reproducing the major features of the experimental isotopic shifts in the dispersion curves. Although a more specific analysis for AF spin modes should be carried out, our analysis suggests that the critical momenta typical of charge modes [stripes and “egg-box”, with $\mathbf{q}_c \sim (\pm\pi/2, 0), (0, \pm\pi/2)$] are an important ingredient to account for ARPES experimental findings.

We acknowledge interesting discussions with C. Castellani, C. Di Castro, G.-H. Gweon, A. Lanzara, who we also thank for showing us her data before publication, and J. Lorenzana. M. G. acknowledges financial support from MIUR Cofin 2003 and G. S. from the Deutsche Forschungsgemeinschaft.

-
- [1] For a review see, e.g., H. Keller, *Unconventional isotope effects in cuprate superconductors*, in *Structure and Bonding*, Springer Verlag, 2004, and references therein.
 - [2] D. Rubio Temprano, Phys. Rev. Lett. **84**, 1990 (2000).
 - [3] G.-H. Gweon, *et al.*, Nature **430**, 187 (2004).
 - [4] E. G. Maksimov, *et al.*, cond-mat/0408251.

- [5] C. Castellani, C. Di Castro, and M. Grilli, Phys. Rev. Lett. **75**, 4650 (1995).
- [6] For a list of early evidences see C. Castellani, *et al.*, J. of Phys. and Chem. of Sol. **59**, 1694 (1998); J.L. Tallon, J.W. Loram, Physica C **349**, 53 (2001).
- [7] C. M. Varma, cond-mat/0312385 and references therein; Y. Dagan, *et al.*, Phys. Rev. Lett. **92**, 167001 (2004); Y. Ando, *et al.*, *ibid.*, 247004 (2004); L. Alff, *et al.*, Nature **422**, 698 (2003).
- [8] C. Howald, *et al.*, Phys. Rev. B **67**, 014533 (2003); M. Vershihin, *et al.*, Science **303**, 1995 (2004); McElroy, *et al.*, cond-mat/0406491.
- [9] S. Andergassen, *et al.*, Phys. Rev. Lett. **87**, 056401 (2001).
- [10] G. Seibold and M. Grilli, Phys. Rev. B **63** 224505 (2001).
- [11] M. Eschrig and M. R. Norman, Phys. Rev. Lett. **85**, 3261 (2000), A. V. Chubukov and M. R. Norman, cond-mat/0402304.
- [12] S. Engelsberg and J. R. Schrieffer, Phys. Rev. **131**, 993 (1963).
- [13] A. Lanzara, *et al.*, Nature **412**, 510 (2001).
- [14] A. Gromko *et al.*, Phys. Rev. B **68**, 174520 (2003).
- [15] G.-H. Gweon, *et al.*, J. Phys. Chem. Solids **65**, 1397 (2004).
- [16] A. P. Kampf and J. R. Schrieffer, Phys. Rev. B **42**, 7967 (1990).
- [17] G. Seibold, *et al.*, Eur. Phys. J. B **13**, 87 (2000).
- [18] J. C. Campuzano, *et al.*, Phys. Rev. Lett. **83**, 3709 (1999).
- [19] A. Kaminski, *et al.*, Phys. Rev. Lett. **86**, 1070 (2001).
- [20] M. R. Norman, Phys. Rev. B **61**, 16117 (2000).
- [21] This finding differs from the result of Ref. [4], where calculations were carried out for an Holstein phonon within a particle-hole symmetric model and keeping (within our notations) the g^2/ω_0 independent on the isotopic mass.
- [22] G. Seibold and M. Grilli, to be published.